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(54) 【発明の名称】 封止用エポキシ樹脂組成物及び電子部品装置

## (57) 【要約】

【課題】レーザーマーク性や電気特性に優れ、パッド間やワイヤー間距離が狭い電子部品装置においても、導電性物質によるショート不良が発生せず、かつ成形性、信頼性、パッケージ表面の外観に優れた封止用エポキシ樹脂組成物及びそれを用いた電子部品装置を提供する。

【解決手段】(A) エポキシ樹脂、(B) 硬化剤、(C) 非導電性カーボン、(D) 無機充填材及び(E) 有機染料を必須成分とし、該(E) 有機染料がアジン系染料、アントラキノン系染料、ジスアゾ系染料、ジイミニウム系染料、アミニウム系染料、ジイモニウム系染料及びフタロシアニン系染料から選ばれる染料を含む封止用エポキシ樹脂組成物、及びこの封止用エポキシ樹脂組成物により封止された素子を備えた電子部品装置。

【選択図】なし

## 【特許請求の範囲】

## 【請求項 1】

(A) エポキシ樹脂、(B) 硬化剤、(C) 非導電性カーボン、(D) 無機充填材及び(E) 有機染料を必須成分とし、該(E) 有機染料がアジン系染料、アントラキノン系染料、ジアゾ系染料、ジイミニウム系染料、アミニウム系染料、ジイモニウム系染料及びフタロシアニン系染料から選ばれる染料を含む封止用エポキシ樹脂組成物。

## 【請求項 2】

(E) 染料がフタロシアニン系染料を含む請求項 1 記載の封止用エポキシ樹脂組成物。

## 【請求項 3】

非導電性カーボンの電気抵抗が  $10^7 \Omega$  以上である請求項 1 または 2 記載の封止用エポキシ樹脂組成物。 10

## 【請求項 4】

非導電性カーボンの含有量が、樹脂組成物全体の 0.1 ~ 1.0 重量% である請求項 1 ~ 3 のいずれか記載の封止用エポキシ樹脂組成物。 15

## 【請求項 5】

非導電性カーボンは、エポキシ樹脂及びフェノール樹脂の少なくともいずれかにより表面が覆われている請求項 1 ~ 4 のいずれか記載の封止用エポキシ樹脂組成物。

## 【請求項 6】

請求項 1 ~ 5 のいずれかに記載の封止用エポキシ樹脂組成物により封止された素子を備えた電子部品装置。 20

## 【発明の詳細な説明】

## 【技術分野】

## 【0001】

本発明は、レーザーマーク性、電気特性、成形性、信頼性及びパッケージ表面の外観に優れた封止用エポキシ樹脂組成物及びこれにより封止された素子を備えた電子部品装置に関する。 25

## 【背景技術】

## 【0002】

I C、L S I 等の半導体素子は、素子の集積度の向上と共に、素子のパッケージに対する占有体積の拡大、半導体装置等の電子部品装置の小型化、薄型化、多ピン化が進んでいく。さらに、電子機器の小型化、薄型化に対応し、実装方法も高密度実装を可能とする表面実装方式がピン挿入方式に代わり、急速に普及している。その結果、電子部品装置を基板へ取り付ける時に、電子部品装置自体が短時間の内に  $200^{\circ}\text{C}$  以上の半田浴といった高温に曝される。この時、封止材中に含まれる水分が気化し、ここで発生する蒸気圧が封止材と素子、リードフレーム等のインサートとの界面において剥離応力として働き、封止材とインサートとの間で剥離が発生し、特に薄型の電子部品装置においては、電子部品装置のフクレやクラックに至ってしまう。 30

このような剥離起因によるフクレ、クラックの防止策として、素子表面又はリードフレームのアイランド裏面にコート材を用いて封止材との密着性を向上させる手法、リードフレームのアイランド裏面にディンプル加工やスリット加工等を行なう、あるいは L O C (Lead on Chip) 構造にして封止材との密着性を向上させる手法が用いられているが、高コスト化、効果が不十分等の問題がある。さらに、L O C 構造の場合、パッケージ表面の色むらが生じ外観を損ねる。 40

また、半導体装置等の電子部品装置の小型化、多ピン化に伴い、インナーリード間やバッド間、ワイヤー間などピッチ間距離が狭くなってきており、封止材中に含まれるカーボンブラック等の導電性物質が大きな粗粒として存在する場合には、インナーリード間やバッド間そしてワイヤー間に導電性であるカーボンブラックが狭まり、電気特性不良に至ってしまう。このため、着色剤としてカーボンブラックの代わりに有機染料や顔料等を用いる検討がなされている（特許文献 1、特許文献 2 参照。）が、YAG レーザーマーク性的低下、信頼性の低下、高コスト化等の問題があった。 50

【特許文献1】特開昭63-179921号公報

【特許文献2】特開平11-60904号公報

【発明の開示】

【発明が解決しようとする課題】

【0003】

本発明は、かかる状況に鑑みてなされたもので、レーザーマーク性や電気特性に優れ、パッド間やワイヤー間距離が狭い半導体装置等の電子部品装置においても、導電性物質によるショート不良が発生せず、かつ成形性、信頼性、パッケージ表面の外観に優れた封止用エポキシ樹脂組成物及びこれにより封止した素子を備えた電子部品装置を提供することを目的とする。

【課題を解決するための手段】

【0004】

本発明者らは上記の課題を解決するために銳意検討を重ねた結果、封止用エポキシ樹脂組成物に非導電性カーボンを配合することにより上記の目的を達成しうることを見い出し、本発明を完成するに至った。

すなわち、本発明は、

(1) (A) エポキシ樹脂、(B) 硬化剤、(C) 非導電性カーボン、(D) 無機充填材及び(E) 有機染料を必須成分とし、該(E) 有機染料がアジン系染料、アントラキノン系染料、ジアゾ系染料、ジミニウム系染料、アミニウム系染料、ジイモニウム系染料及びフタロシアニン系染料から選ばれる染料を含む封止用エポキシ樹脂組成物、

(2) (E) 染料がフタロシアニン系染料を含む上記(1)記載の封止用エポキシ樹脂組成物、

(3) 非導電性カーボンの電気抵抗が $10^7\Omega$ 以上である上記(1)または(2)記載の封止用エポキシ樹脂組成物、

(4) 非導電性カーボンの含有量が、樹脂組成物全体の0.1～10重量%である上記(1)～(3)のいずれか記載の封止用エポキシ樹脂組成物、

(5) 非導電性カーボンは、エポキシ樹脂及びフェノール樹脂の少なくともいずれかにより表面が覆われている上記(1)～(4)のいずれか記載の封止用エポキシ樹脂組成物、及び

(6) 上記(1)～(5)のいずれかに記載の封止用エポキシ樹脂組成物により封止された素子を備えた電子部品装置

に関する。

【発明の効果】

【0005】

本発明による封止用エポキシ樹脂組成物は、実施例に示したように成形性が良好で、レーザーマーク性、電気特性、信頼性、パッケージ表面の外観に優れる電子部品装置が得られるので、その工業的価値は大である。

【発明を実施するための最良の形態】

【0006】

本発明において用いられる(A)エポキシ樹脂としては特に制限はないが、例えば、封止用エポキシ樹脂組成物で一般に使用されている、フェノールノボラック型エポキシ樹脂、オルソクレゾールノボラック型エポキシ樹脂、ビスフェノールAノボラック型エポキシ樹脂をはじめとするフェノール類とアルデヒド類から合成されるノボラック樹脂をエポキシ化したエポキシ樹脂、アルキル置換又は非置換のビフェノール型エポキシ樹脂、アルキル置換又は非置換のグリシジルエーテル型エポキシ樹脂、イソシアヌル酸等のポリアミンとエピクロロヒドリンの反応により得られるグリシジルアミン型エポキシ樹脂、ビフェニル型エポキシ樹脂、ジシクロベンタジエンとフェノール類及び/又はナフトール類の共縮合樹脂のエポキシ化物、ナフタレン環を有するエポキシ樹脂、フェノール・アラルキル樹脂、ナフトール・アラルキル樹脂等のアラルキル型フェノール樹脂のエポキシ化物、トリメチロールプロパン型エポキシ樹脂、テルペン変性エポキシ樹脂、オレフィン結合を過酢

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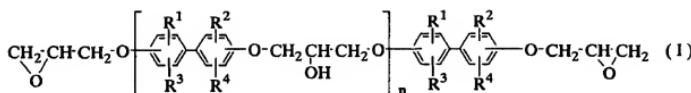
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酸等の過酸で酸化して得られる線状脂肪族エポキシ樹脂、脂環族エポキシ樹脂などが挙げられ、これらは単独で用いても2種以上を組み合わせて用いてもよい。

## 【0007】

具体的には、下記一般式(I)～(IV)のいずれかにより示される化合物などが挙げられる。

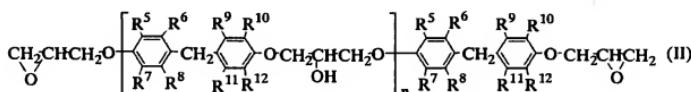
## 【化1】



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(ここで、R<sup>1</sup>～R<sup>4</sup>は水素原子及び炭素数1～10の置換又は非置換の一価の炭化水素基から選ばれ、全てが同一でも異なっていてもよい。nは0～3の整数を示す。)

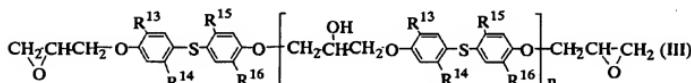
## 【化2】



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(ここで、R<sup>5</sup>～R<sup>12</sup>は水素原子及び炭素数1～10の置換又は非置換の一価の炭化水素基から選ばれ、全てが同一でも異なっていてもよい。nは0～3の整数を示す。)

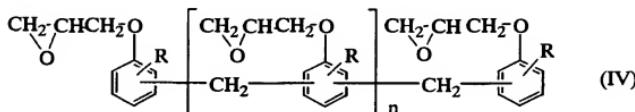
## 【化3】



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(ここで、R<sup>13</sup>～R<sup>16</sup>は水素及び炭素数1～10の置換又は非置換の一価の炭化水素基から選ばれ、すべて同一でも異なっていてもよい。nは0～10の整数を示す。)

## 【化4】



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(ここで、Rは水素及び炭素数1～10の置換又は非置換の一価の炭化水素基から選ばれ、nは0～10の整数を示す。)

## 【0008】

上記一般式(I)中のR<sup>1</sup>～R<sup>4</sup>は、水素原子、メチル基、エチル基、プロピル基、ブ

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チル基、イソブロピル基、イソブチル基等のアルキル基、メトキシ基、エトキシ基、プロポキシ基、ブトキシ基等のアルコキシル基、フェニル基、トリル基、キシリル基等のアリール基、ベンジル基、フェネチル基等のアラルキル基、メトキシメチル基、エトキシメチル基メトキシエチル基等のアルコキシアルキル基、アミノメチル基、アミノエチル基等のアミノ基置換アルキル基などの炭素数1～10の置換又は非置換の一価の炭化水素基から選ばれるが、中でも水素原子又はメチル基が好ましい。上記一般式(1)で示されるビフェニル型エポキシ樹脂としては、4, 4'-ビス(2, 3-エポキシプロポキシ)ビフェニル又は4, 4'-ビピス(2, 3-エポキシプロポキシ)-3, 3', 5, 5'-テトラメチルビフェニルを主成分とするエポキシ樹脂、エピクロロヒドリンと4, 4'-ビフェノール又は4, 4'-(3, 3', 5, 5'-テトラメチル)ビフェノールとを反応させて得られるエポキシ樹脂等が挙げられ、中でも、4, 4'-ビス(2, 3-エポキシプロポキシ)-3, 3', 5, 5'-テトラメチルビフェニルを主成分とするエポキシ樹脂がより好ましい。

## 【0009】

上記一般式(II)中のR<sup>5</sup>～R<sup>12</sup>は、水素原子、メチル基、エチル基、プロピル基、ブチル基、イソブロピル基、イソブチル基等のアルキル基、メトキシ基、エトキシ基、プロポキシ基、ブトキシ基等のアルコキシル基、フェニル基、トリル基、キシリル基等のアリール基、ベンジル基、フェネチル基等のアラルキル基、メトキシメチル基、エトキシメチル基メトキシエチル基等のアルコキシアルキル基、アミノメチル基、アミノエチル基等のアミノ基置換アルキル基などの炭素数1～10の置換又は非置換の一価の炭化水素基から選ばれるが、中でも水素原子又はメチル基が好ましい。上記一般式(II)で示されるビスフェノールF型エポキシ樹脂としては、例えば、R<sup>5</sup>、R<sup>7</sup>、R<sup>10</sup>及びR<sup>12</sup>がメチル基で、R<sup>6</sup>、R<sup>8</sup>、R<sup>9</sup>及びR<sup>11</sup>が水素原子であり、n=0を主成分とするE S L V-80XY(新日鉄化学株式会社製商品名)が市販品として入手可能である。

## 【0010】

上記一般式(III)中のR<sup>13</sup>～R<sup>16</sup>は、水素原子、メチル基、エチル基、プロピル基、イソブロピル基、n-ブチル基、t-ブチル基、アミル基、イソブチル等のアルキル基、メトキシ基、エトキシ基、プロポキシ基、ブトキシ基等のアルコキシル基、フェニル基、トリル基、キシリル基等のアリール基、ベンジル基、フェネチル基等のアラルキル基、メトキシメチル基メトキシエチル基等のアルコキシアルキル基、アミノメチル基、アミノエチル基等のアミノ基置換アルキル基などの炭素数1～10の置換又は非置換の一価の炭化水素基から選ばれるが、中でもアルキル基が好ましく、メチル基又はt-ブチル基がより好ましい。上記一般式(III)で示されるフェニルスルフィド型エポキシ樹脂としては、入手性及び流動性的観点から、R<sup>13</sup>及びR<sup>16</sup>がt-ブチル基で、R<sup>14</sup>及びR<sup>15</sup>がメチル基で、n=0である化合物を主成分とするビス(2-メチル-4-ヒドロキシ-5-t-ブチル)チオエーテルのエポキシ化物が好ましい。

## 【0011】

上記一般式(IV)中のRとしては、例えば、水素、メチル基、エチル基、プロピル基、ブチル基、イソブロピル基、t-ブチル基等のアルキル基、ビニル基、アリル基、ブテニル基等のアルケニル基、ハロゲン化アルキル基、アミノ基置換アルキル基、メルカプト基置換アルキル基などの炭素数1～10の置換又は非置換の一価の炭化水素基が挙げられ、中でもメチル基、エチル基等のアルキル基が好ましく、メチル基がより好ましい。上記一般式(IV)で示されるノボラック型エポキシ樹脂としては、フェノールノボラック型エポキシ樹脂、オルソクレゾールノボラック型エポキシ樹脂等が挙げられ、中でもオルソクレゾールノボラック型エポキシ樹脂が好ましい。

## 【0012】

本発明において用いられる(B)硬化剤としては特に制限はないが、例えば、封止用エポキシ樹脂成物で一般に使用されているもので、フェノール、クレゾール、レゾルシン、カテコール、ビスフェノールA、ビスフェノールF等のフェノール類及び/又はα-ナフトール、β-ナフトール、ジヒドロキシナフタレン等のナフトール類とホルムアルデヒ

ド等のアルデヒド類とを酸性触媒下で縮合又は共縮合させて得られる樹脂、フェノール・アラルキル樹脂、ナフトル・アラルキル樹脂等のアラルキル型フェノール樹脂などが挙げられ、これらは単独で用いても2種以上を組み合わせて用いてもよい。

(B) 硬化剤は、上記(A)エポキシ樹脂のエポキシ基当量に対して、フェノール性水酸基当量が0.5～1.5当量となる量で配合されることが好ましく、より好ましくは0.8～1.2当量で配合される。0.5当量未満ではエポキシ樹脂の硬化が不十分となり、硬化物の耐熱性、耐湿性並びに電気特性が劣る傾向があり、1.5当量を超えると硬化剤成分が過剰になり硬化樹脂中に多量のフェノール性水酸基が残るため、電気特性並びに耐湿性が悪くなる傾向がある。

## 【0013】

本発明の封止用エポキシ樹脂組成物には、エポキシ基とフェノール性水酸基とのエーテル化反応を促進するため、硬化促進剤を配合することが好ましい。硬化促進剤としては、エポキシ樹脂と硬化剤の反応を促進させるような触媒機能を持つ化合物であれば特に制限はないが、例えば、1,8-ジアザビシクロ(5,4,0)ウンデセン-7、1,5-ジアザビシクロ(4,3,0)ノネン、5,6-ジチルアミノ-1,8-ジアザビシクロ(5,4,0)ウンデセン-7、ベンジルジメチルアミン、トリエタノールアミン、ジメチルアミノエタノール、トリス(ジメチルアミノメチル)フェノール等の3級アミン類及びこれらの誘導体、2-メチルイミダゾール、2-フェニルイミダゾール、2-フェニル-4-メチルイミダゾール等のイミダゾール類及びこれらの誘導体、トリプチルホスフィン、メチルジフェニルホスフィン、トリフェニルホスフィン、ジフェニルホスフィン、フェニルホスフィン等の有機ホスフィン類及びこれらのホスフィン類に無水マレイン酸、ベンゾキノン、ジアゾフェニルメタン等の $\alpha$ 結合をもつ化合物を付加してなる分子内分極を有するリン化合物、テトラフェニルホスホニウムテトラフェニルボレート、トリフェニルホスフィンテトラフェニルボレート、2-エチル-4-メチルイミダゾールテトラフェニルボレート、N-メチルテトラフェニルホスホニウムテトラフェニルボレート、トリフェニルホスホニウム-トリフォニルボラン等が挙げられ、これらは単独で用いても2種以上を組み合わせて用いてもよい。

硬化促進剤の配合量は、エポキシ樹脂100重量部に対して0.01～5重量部が好ましく、0.1～3重量部を配合するのがより好ましい。0.01重量部より少ないと促進効果が小さく、5重量部より多いと保存安定性が悪くなる傾向にある。

## 【0014】

本発明において用いられる(C)非導電性カーボンとしては特に制限はないが、例えばポリマーを焼成させて得られる非導電性ポリマー焼成カーボン、カーボンブラックの表面にポリマーをグラフトさせて得られるグラフトカーボン、カーボンブラックの表面をシリカ等の絶縁無機物で覆うことによって得られるカーボン内包フィラー、酸化処理等の表面処理を施したカーボンブラックなどが挙げられ、これらは単独で用いても2種以上を組み合わせて用いてもよい。また、これらの非導電性カーボンをエポキシ樹脂、フェノール樹脂等でコートして用いることもできる。

非導電性ポリマー焼成カーボンの製造法には特に制限はないが、例えば、ナフタレン等の芳香族を有するスルホン酸ポリマーなどのポリマーを400～600℃で焼成する方法が挙げられる。

グラフトカーボンの製造法には特に制限はないが、例えば、エポキシ基、チオエポキシ基、アジリジン基、オキサゾリン基、イソシアネット基等のカーボンブラック表面のカルボキシル基と反応性を有する基を有するグラフトポリマーを、熱によりカーボンブラックに不可逆的付加反応させる方法が挙げられる。ここで用いられるグラフトポリマーとしては特に制限はないが、上記の反応性基を有する不飽和化合物をそれと共に重合可能な单量体と共に重合させて得られるもので、例えば、ポリスチレン、ポリエチレングリコール、ポリ

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オキサゾリン、ポリプロピレンジリコール、ポリメタクリル酸メチル、アクリル樹脂、フッ素樹脂等及びこれららの誘導体、ポリジメチルシリコキサン等のシリコーン、ワックスなどが挙げられ、これららは単独で用いても2種以上を組み合わせて用いてもよい。グラフトカーボンに用いられるカーボンブラックとしては特に制限はないが、グラフトポリマーとの反応点であるカルボキシル基等の酸性官能基が表面に多く存在するものが好ましく、pHが7以下のカーボンブラックがより好ましい。グラフトカーボンのグラフト率は非導電性の観点から30重量%以上が好ましく、50重量%以上がより好ましい。ここで、グラフト率とはグラフトカーボン全体に占めるグラフトポリマーの割合をさす。

カーボン内包フィラーの製造法には特に制限はないが、例えば、水溶液中に分散させたカーボンブラックの表面に、テトラエキシシランを加水分解させることによってシリカ微粒子を沈着させる方法、水溶液中に分散させたカーボンブラックにシリカゾルを添加しゾルゲル法によってシリカ微粒子を沈着させる方法等が挙げられる。カーボンブラックの表面を覆う絶縁無機物としては特に制限はないが、例えば、炭酸カルシウム、炭酸バリウム等のアルカリ土類金属炭酸塩、珪酸カルシウム、珪酸マグネシウム等のアルカリ土類金属珪酸塩、酸化鉄、シリカ、アルミナ、酸化銅、酸化コバルト、酸化ニッケル等の金属酸化物、炭酸コバルト、炭酸ニッケル、塩基性炭酸銅、水酸化アルミニウム、ダイレクトブラック154カルシウムレーキなどが挙げられ、これららは単独で用いても2種以上を組み合わせて用いてもよい。カーボン内包フィラーに用いられるカーボンブラックとしては特に制限はないが、水に濡れやすく、かさ高くないものが好ましい。カーボン内包フィラーのカーボン内包率は、非導電性の観点から70重量%以下が好ましく、50重量%以下がより好ましい。ここで、カーボン内包率とは、カーボン内包フィラー全体に占めるカーボンブラックの割合をさす。

表面処理を施したカーボンブラックの製造法としては、特に制限はないが、例えば、空気酸化法、硝酸、窒素酸化物と空気の混合ガス、オゾン等の酸化剤を用いる酸化処理法、シランカップリング剤を用いる方法等でカーボンブラックを表面処理する方法などが挙げられる。

#### 【0015】

(C) 非導電性カーボンの電気抵抗は、 $10^7\ \Omega$ 以上が好ましく、 $10^9\ \Omega$ 以上がより好ましい。電気抵抗は、JIS Z 3197に準拠した、ガラス布基材エポキシ樹脂鋼張積層板MCL-E67(日立化成工業株式会社製商品名)を基板とした2形のくし型電極(銅箔厚3.5μm、線幅0.3mm、線間0.3mm、基板厚0.95mm)を用い、粉末サンプル0.5gを配線上にセロハンテープで固定し、抵抗計(タケダ理研工業株式会社製TR8601)により直流500Vで1分後の抵抗値を測定することにより求められる。

(C) 非導電性カーボンの平均粒径は、0.3~50μmが好ましく、0.3~30μmがより好ましい。レーザーマーク性、成形性の観点からは0.3~10μmが好ましく、0.3~5μmがより好ましい。

(C) 非導電性カーボンの含有量は、封止用エポキシ樹脂組成物全体に対し0.1~10重量%が好ましく、0.3~8重量%がより好ましく、0.5~4重量%がさらに好ましい。0.1重量%未満ではパッケージ表面の外観が損なわれたり、遮光性、レーザーマーク性が不十分となったりする場合があり、10重量%を超えると成形性が不十分となる傾向がある。

#### 【0016】

充填材としては、吸湿性低減及び強度向上の観点から無機充填材を用いることが必要である。本発明に用いられる(D)無機充填材としては特に制限はないが、例えば、封止用エポキシ樹脂組成物で一般に使用されているもので、溶融シリカ、結晶シリカ、アルミナ、ジルコン、珪酸カルシウム、炭酸カルシウム、炭化珪素、窒化ホウ素、ベリリア、ジルコニア等の粉体、又はこれらを球形化したビーズ、チタン酸カリウム、炭化珪素、窒化珪素、アルミナ等の単結晶繊維、ガラス繊維などが挙げられ、これららは単独で用いても2種以上を組み合わせて用いてもよい。また、難燃効果のある無機充填材としては水酸化アル

ミニウム、鋼機亜鉛等が挙げられ、これらは単独で用いても併用してもよい。中でも、線膨張係数低減の観点からは溶融シリカが、高熱伝導性の観点からはアルミナが好ましく、無機充填材の形状は、成形時の流動性及び金型摩耗性の点から球形が好ましい。

(D) 無機充填材の配合量としては、吸湿性、線膨張係数の低減、強度向上及び半田耐熱性の観点から、封止用エポキシ樹脂組成物全体に対し70～98重量%が好ましい。より好ましくは、75～95重量%の範囲である。

#### 【0017】

本発明の封止用エポキシ樹脂組成物には、無機充填材と樹脂成分との親和性をはかる観点からはカップリング剤を添加することが好ましい。用いられるカップリング剤としては特に制限はなく、例えば、封止用エポキシ樹脂組成物で一般に使用されているもので、ビニルトリクロロシラン、ビニルトリエトキシシラン、ビニルトリス(β-メトキシエトキシ)シラン、γ-メタクリロキシプロピルトリメトキシシラン、β-(3,4-エポキシジクロロヘキシル)エチルトリメトキシシラン、γ-グリシドキシプロピルトリメトキシシラン、ビニルトリエトキシシラン、γ-メルカプトプロピルトリメトキシシラン、γ-アミノブロピルトリエトキシシラン、γ-[ビス(β-ヒドロキシエチル)]アミノブロピルトリエトキシシラン、N-β-(アミノエチル)-γ-アミノブロピルトリメトキシシラン、γ-(β-アミノエチル)アミノブロピルジメトキシメチルシラン、N-(トリメトキシリルブロピル)エチレンジアミン、N-(ジメトキシメチルシリルイソプロピル)エチレンジアミン、メチルトリメトキシシラン、メチルトリエトキシシラン、N-β-(N-ビニルベンジルアミノエチル)-γ-アミノブロピルトリメトキシシラン、γ-エクロブロピルトリメトキシシラン、ヘキサメチルジシラン、γ-アミニノブロピルトリメトキシシラン、ビニルトリメトキシシラン、γ-メルカプトブロピルメチルジメトキシシラン等のシラン系カップリング剤、イソブロピルトリイソステアロイルチタネット、イソブロピルトリス(ジオクチルバイオホスフェート)チタネット、イソブロピルトリ(N-アミノエチルアミノエチル)チタネット、テトラオクチルビス(ジトリデシルホスファイト)チタネット、テトラ(2,2-ジアリルオキシメチル-1-ブチル)ビス(ジトリデシル)ホスファイトチタネット、ビス(ジオクチルバイオホスフェート)オキシアセテートチタネット、ビス(ジオクチルバイオホスフェート)エチレンチタネット、イソブロピルトリオクタノイルチタネット、イソブロピルジメタクリルイソステアロイルチタネット、イソブロピルトリドデシルベンゼンスルホニルチタネット、イソブロピルイソステアロイルジアクリルチタネット、イソブロピルトリ(ジオクチルホスフェート)チタネット、イソブロピルトリクミルフェニルチタネット、テトライソブロピルビス(ジオクチルホスファイト)チタネット等のチタネット系カップリング剤などが挙げられ、これらは単独で用いても2種以上を組み合わせて用いてもよい。

#### 【0018】

その他の添加剤として、高級脂肪酸例えはカルナバワックスとポリエチレン系ワックス等の離型材、シリコーンオイル、シリコーンゴム等の改質材、ハイドロタルサイト、アンチモン-ビスマス等のオイントラッパーなどを必要に応じて、それぞれ単独で又は2種以上を組み合わせて配合することができる。また、本発明の封止用エポキシ樹脂組成物には、本発明の効果が達成される範囲内であれば、非導電性カーボン以外に、アジン系染料、アントラキノン系染料、ジスアゾ系染料、ジイミニウム系染料、ジミニウム系染料、ジイモニウム系染料、C<sub>r</sub>錯体、F<sub>e</sub>錯体、C<sub>o</sub>錯体、N<sub>i</sub>錯体、F<sub>e</sub>、C<sub>u</sub>、N<sub>i</sub>等の金属化合物、A<sub>l</sub>、M<sub>g</sub>、F<sub>e</sub>等の金属酸化物、雲母、近赤外吸收材、フタロシアニン系顔料、フタロシアニン系染料、カーボンブラックなどの着色剤を1種又は2種以上併用することができる。特に、レーザーマーク性、流動性、硬化性の点からはフタロシアニン系染料を併用することが好ましい。カーボンブラック等の導電性粒子を併用する場合には、本発明の目的を達成するために、分散性に優れる、比表面積が130m<sup>2</sup>/g以下、D B P吸油量が120cm<sup>3</sup>/100g以下のものが好ましく、比表面積が100m<sup>2</sup>/g以下、D B P吸油量が80cm<sup>3</sup>/100g以下のものがより好ましい。

#### 【0019】

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本発明の封止用エポキシ樹脂組成物は、上記各原材料を均一に分散混合できるのであれば、いかなる手法を用いても調製できるが、一般的な方法としては、所定の配合量の原材料をミキサー等によって充分混合した後、熱ロール、押出機等によって混練し、冷却、粉碎する方法を挙げることができる。成形条件に合うような寸法及び重量でタブレット化すると使いやすい。

#### 【0020】

本発明の封止用エポキシ樹脂組成物は、レーザーマーク性、電気特性に優れ、且つ成形性、信頼性、スケルトン防止に優れており、I C、L S I 等の封止に好適に用いることができる。

本発明で得られる封止用エポキシ樹脂組成物により封止した素子を備えた電子部品装置としては、リードフレーム、配線済みのテープキャリア、配線板、ガラス、シリコンウエハ等の支持部材に、半導体チップ、トランジスタ、ダイオード、サイリスタ等の能動素子、コンデンサ、抵抗体、コイル等の受動素子等の素子を搭載し、必要な部分を本発明の封止用エポキシ樹脂組成物で封止した、電子部品装置などが挙げられる。このような電子部品装置としては、例えば、リードフレーム上に半導体素子を固定し、ポンディングパッド等の素子の端子部とリード部をワイヤポンディングやバンプで接続した後、本発明の封止用エポキシ樹脂組成物を用いてトランシスファ成形などにより封止してなる、D I P (Dual Inline Package)、P L C C (Plastic Leaded Chip Carrier)、Q F P (Quad Flat Package)、S O P (Small Outline Package)、S O J (Small Outline J-lead package)、T S O P (Thin Small Outline Package)、T Q F P (Thin Quad Flat Package) 等の一般的な樹脂封止型I C、テープキャリアにバンプで接続した半導体チップを、本発明の封止用エポキシ樹脂組成物で封止したT C P (Tape Carrier Package)、配線板やガラス上に形成した配線に、ワイヤポンディング、フリップチップポンディング、はんだ等で接続した半導体チップ、トランジスタ、ダイオード、サイリスタ等の能動素子及び／又はコンデンサ、抵抗体、コイル等の受動素子を、本発明の封止用エポキシ樹脂組成物で封止したC O B (Chip On Board) モジュール、ハイブリッドI C、マルチチップモジュール、裏面に配線板接続用の端子を形成した有機基板の表面に素子を搭載し、バンプまたはワイヤポンディングにより素子と有機基板に形成された配線を接続した後、本発明の封止用エポキシ樹脂組成物で素子を封止したB G A (Ball Grid Array)、C S P (Chip Size Package) などが挙げられる。また、プリント回路板にも本発明の封止用エポキシ樹脂組成物は有效地に使用できる。

本発明で得られる封止用エポキシ樹脂組成物を用いて電子部品装置を封止する方法としては、低圧トランシスファ成形法が最も一般的であるが、インジェクション成形、圧縮成形、注型等の方法を用いてもよい。

#### 【実施例】

#### 【0021】

以下に本発明を実施例により詳細に説明するが、本発明はこれに限定されるものではない。

実施例1～9、比較例1～4

まず、表1及び表2に示す配合成分を予備混合（ドライブレンド）した後、二軸ロール（ロール表面温度80°C）で10分間混練し、冷却粉碎して実施例及び比較例の封止用エポキシ樹脂組成物を製造した。

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【表1】

表1 配合組成1

(配合量:重量部)

配合成分	実施例								
	1	2	3	4	5	6	7	8	9
ビフェニル型エポキシ樹脂	85.0	85.0	85.0	85.0	85.0	85.0	85.0	85.0	85.0
臭素化エポキシ樹脂	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0
アラルキル型フェノール樹脂	87.0	87.0	87.0	87.0	87.0	87.0	87.0	87.0	87.0
トリフェニルホスフィン	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
エポキシシラン	7.6	7.6	7.6	7.6	7.6	7.6	7.6	7.6	7.6
カルナバワックス	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
ポリエチレンワックス	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
非導電性カーボンA	55.0	-	-	20.0	20.0	10.0	-	-	30.0
非導電性カーボンB	-	60.0	-	-	30.0	20.0	-	-	-
非導電性カーボンC	-	-	65.0	-	-	30.0	-	-	-
非導電性カーボンD	-	-	-	-	-	-	50.0	-	-
非導電性カーボンE	-	-	-	-	-	-	-	30.0	-
カーボンブラックA	-	-	-	2.0	-	-	-	-	-
フタロシアニン系染料	-	-	-	-	-	-	-	-	1.0
三酸化アンチモン	12	12	12	12	12	12	12	12	12
溶融シリカ	1550	1550	1550	1550	1550	1550	1550	1550	1550

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【0023】

【表2】

表2 配合組成2

(配合量:重量部)

配合成分	比較例			
	1	2	3	4
ビフェニル型エポキシ樹脂	85.0	85.0	85.0	85.0
臭素化エポキシ樹脂	15.0	15.0	15.0	15.0
アラルキル型フェノール樹脂	87.0	87.0	87.0	87.0
トリフェニルホスフィン	3.0	3.0	3.0	3.0
エポキシシラン	7.6	7.6	7.6	7.6
カルナバワックス	1.5	1.5	1.5	1.5
ポリエチレンワックス	1.5	1.5	1.5	0.5
カーボンブラックB	3.5	1.5	-	-
アジン系染料	-	10.0	35.0	-
雲母-無酸化鉄系染料	-	-	-	30.0
三酸化アンチモン	12	12	12	15
溶融シリカ	1550	1550	1550	1550

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【0024】

なお、表1及び表2記載の配合成分は次の通りである。

ビフェニル型エポキシ樹脂：油化シェルエポキシ株式会社製商品名YX-4000H

臭素化エポキシ樹脂：東都化成株式会社製商品名YDB-400

アラルキル型フェノール樹脂：三井化学株式会社製商品名XL-225-3L

エポキシシラン：日本ユニカー株式会社製商品名A-187

ポリエチレンワックス：クラリアントジャパン株式会社製商品名PED-191

非導電性カーボンA：三井鉱山株式会社製商品名CB-3-500（平均粒径3μm）、

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電気抵抗  $10^9 \Omega$ )

非導電性カーボン B : 三井鉱山株式会社製商品名 C B - 7 - 5 0 0 (平均粒径  $7 \mu\text{m}$ 、電気抵抗  $10^9 \Omega$ )

非導電性カーボン C : 三井鉱山株式会社製商品名 C B - 1 0 - 5 0 0 (平均粒径  $10 \mu\text{m}$ 、電気抵抗  $10^9 \Omega$ )

非導電性カーボン D : スチレン／アクリル系グラフトポリマーを用いたグラフト率 90 重量% のグラフトカーボン (株式会社日本触媒製、電気抵抗  $10^{10} \Omega$ )

非導電性カーボン E : カーボン内包率 17 重量% のカーボン内包シリカ (鈴木油脂工業株式会社製、平均粒径  $7 \mu\text{m}$ 、電気抵抗  $10^9 \Omega$ )

カーボンブラック A : 比表面積  $8.9 \text{ m}^2/\text{g}$ 、D B P 吸油量  $60 \text{ cm}^3/100 \text{ g}$  のカーボンブラック

カーボンブラック B : 比表面積  $14.0 \text{ m}^2/\text{g}$ 、D B P 吸油量  $131 \text{ cm}^3/100 \text{ g}$  のカーボンブラック

フタロシアニン系染料 : 山本化成株式会社製商品名 Y K R - 3 0 8 0

アジン系染料 : 住友化学工業株式会社製商品名スピリットブラック 9 2 0

雲母－黒酸化鉄系染料 : メルクジャパン株式会社製商品名 L S 8 3 5

溶融シリカ : マイクロン株式会社製商品名 S - C O

【0025】

実施例、比較例の封止用エポキシ樹脂組成物を次に示す各試験により評価した。なお、封止用エポキシ樹脂組成物の成形は、トランスファ成形機を用い、金型温度  $180^\circ\text{C}$ 、成形圧力  $6.9 \text{ MPa}$ 、硬化時間 90 秒の条件で行なった。また、後硬化 (ポストキュア) は  $175^\circ\text{C}$  で 6 時間行った。

#### (1) スパイラルフロー

E M M 1 1 - 6 6 に準じた金型をトランスファ成形機にセットし、上記条件で封止用エポキシ樹脂組成物を成形し、流動距離 ( $\text{cm}$ ) を求めた。

#### (2) 热時硬度

バリ測定金型 (幅  $5 \text{ mm}$ 、深さ  $50$ 、 $30$ 、 $20$ 、 $10$ 、 $5$ 、 $2 \mu\text{m}$  のスリットを設けた金型) をトランスファ成形機にセットし、上記条件で封止用エポキシ樹脂組成物を成形し、金型開放 0 秒後、樹脂滴り部分をショア硬度計にて測定した。

#### (3) 体積抵抗率

円板金型をトランスファ成形機にセットし、封止用エポキシ樹脂組成物を上記条件で直径  $100 \text{ mm}$ 、厚さ  $3 \text{ mm}$  の円板に成形し後硬化した後、体積抵抗計を用いて、電圧  $50 \text{ V}$ 、 $150^\circ\text{C}$  で測定し、絶縁性を確認した。

#### (4) 耐湿性

耐湿性に用いた電子部品装置は S O P - 2 8 ピンの S O P 2 8 ピンの樹脂封止型半導体装置 (外形寸法  $1.8 \times 8.4 \times 2.6 \text{ mm}$ ) であり、リードフレームは 42 アロイ材 (加工ディンプル) で  $9.6 \times 5.1 \text{ mm}$  の T E G チップ (A 1 配線  $\pm 0$  及び  $\pm 20 \text{ ミクロン幅}$ 、ギャップ 10 及び  $\pm 20 \text{ ミクロン}$ 、バッショーションなし) を有するもので、上記条件でトランスファ成形、後硬化を行って作製した。 $85^\circ\text{C}/85 \text{ RH\%}$  で 72 時間吸湿した後、 $240^\circ\text{C}/10 \text{ 秒}$  (IR リフロー) の前処理後、P C T 处理 ( $121^\circ\text{C}/0.2 \text{ MPa}$ ) してチップ上配線の断線の有無を確認し、断線パッケージが試験パッケージ中 50% に達するまでの P C T 处理時間で評価した。

#### (5) 半田耐熱性

半田耐熱性に用いた電子部品装置は、Q F P - 8 0 ピンの樹脂封止型半導体装置 (外形寸法  $2.0 \times 1.4 \times 2.0 \text{ mm}$ ) であり、リードフレームは 42 アロイ材 (加工なし) で  $8 \times 10 \text{ mm}$  のチップサイズを有するもので、上記条件でトランスファ成形、後硬化を行つて作製した。

このようにして得られた樹脂封止型半導体装置を、 $85^\circ\text{C}/85 \text{ RH\%}$  で所定の時間吸湿した後、 $240^\circ\text{C}/10 \text{ 秒}$  の処理を行なった時のクラック発生を観察し、外観クラックが発生するまでの吸湿時間により半田耐熱性を評価した。

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## (6) レーザーマーク性

レーザーマーク性評価に用いた電子部品装置は、QFP-54ピンの樹脂封止型半導体装置であり、パッケージ表面をYAGレーザーマーキング装置で、YAGレーザー波長1064nm、レーザーパワー5Jの条件で印字し、目視でマーク性を評価した。

## (7) 電気特性

電気特性の評価に用いた電子部品装置は、LQFP( Lowprofile Quad Flat Package) 176ピンの樹脂封止型半導体装置であり、リーク電流の有無で評価した。

## (8) 黒色度

黒色度は、円板金型をトランスマルチ成形機にセットし、封止用エポキシ樹脂組成物を上記条件で表面が梨地である直径100mm、厚さ2mmの円板に成形し後硬化した後、色差計にて測定した。黒色度は値が小さいほど黒色を示す。

上記の試験結果を表3及び表4に示す。

【0026】

【表3】

表3 評価結果1

項目	単位	実施例								
		1	2	3	4	5	6	7	8	9
スパイラルフロー	cm	84	89	94	99	97	94	81	84	94
熱時硬度	-	80	80	81	80	81	81	80	81	81
体積抵抗率	Ω·cm	$6 \times 10^{13}$	$7 \times 10^{13}$	$6 \times 10^{13}$	$1 \times 10^{13}$	$5 \times 10^{13}$	$4 \times 10^{13}$	$\times 10^{13}$	$5 \times 10^{13}$	$4 \times 10^{13}$
耐湿性	h	650	650	600	720	600	650	650	600	650
半田耐熱性	h	120	120	120	120	120	120	120	120	120
レーザーマーク性	-	良好	良好	良好	良好	良好	良好	良好	良好	良好
電気特性(リーク電流の有無)	-	無	無	無	無	無	無	無	無	無
黒色度	-	26.4	28.9	29.5	26.5	27.6	28.7	28.2	28.3	27.9
総合判定	-	○	○	○	○	○	○	○	○	○

【0027】

【表4】

表4 評価結果2

項目	単位	比較例			
		1	2	3	4
スパイラルフロー	cm	97	89	64	76
熱時硬度	-	81	80	79	80
体積抵抗率	Ω·cm	$3 \times 10^{13}$	$9 \times 10^{11}$	$8 \times 10^{11}$	$8 \times 10^{10}$
耐湿性	h	650	310	240	600
半田耐熱性	h	120	96	48	96
レーザーマーク性	-	良好	良好	良好	良好
電気特性(リーク電流の有無)	-	有	無	無	有
黒色度	-	27.8	28.2	28.5	28.2
総合判定	-	×	×	×	×

【0028】

本発明の着色剤として非導電性カーボンを用いていない比較例1～4は、いずれも満足な特性が得られない。すなわち、導電性のカーボンブラックを用いた比較例1は電気特性に劣り、アジン系染料を用いた比較例2、3は耐湿性及び半田耐熱性に劣る。雲母-黒酸化鉄系染料を用いた比較例4は半田耐熱性及び電気特性に劣る。

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これに対して、実施例1～9は、耐湿性、半田耐熱性等の信頼性及び電気特性に優れ、レーザーマーク性、黒色度も良好である。

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## (54) EPOXY RESIN COMPOSITION FOR SEALING AND ELECTRONIC PART DEVICE

### (57)Abstract:

PROBLEM TO BE SOLVED: To obtain an epoxy resin composition for sealing, excellent in laser marking property and electric characteristics, causing no short circuit due to electroconductive materials even in an electronic part device with a narrow interval between pads or wires and having excellent moldability, reliability and package surface externals, and to provide electronic part devices using the epoxy resin composition.

SOLUTION: The epoxy resin composition for sealing comprises, as essential ingredients, (A) an epoxy resin, (B) a curing agent, (C) non-electroconductive carbon, (D) an inorganic filler and (E) an organic dye, wherein the organic dye includes a dye selected from azine dyes, anthraquinone dyes, disazo dyes, diiminium dyes, aminium dyes, diimmonium dyes and phthalocyanine dyes. The electronic part device is equipped with elements sealed with the epoxy resin composition.

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## CLAIMS

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[Claim(s)]

[Claim 1]

(A) The epoxy resin constituent for the closures containing the color as which an epoxy resin, the (B) curing agent, (C) non-conductive carbon, (D) inorganic filler, and (E) organic dye are used as an indispensable component, and \*\* (E) organic dye is chosen from an azine system color, anthraquinone dye, a JISUAZO system color, a G minium system color, an aminium system color, a G MONIUM system color, and phthalocyanine dye.

[Claim 2]

(E) The epoxy resin constituent for the closures according to claim 1 with which a color contains phthalocyanine dye.

[Claim 3]

The epoxy resin constituent for the closures according to claim 1 or 2 whose electric resistance of non-conductive carbon is 107ohms or more.

[Claim 4]

Claims 1-3 whose contents of non-conductive carbon are 0.1 - 10% of the weight of the whole resin constituent are the epoxy resin constituents for the closures of a publication either.

[Claim 5]

For non-conductive carbon, claims 1-4 of an epoxy resin and phenol resin by which the front face is covered with either at least are the epoxy resin constituents for the closures of a publication either.

[Claim 6]

Electronic-parts equipment equipped with the component by which the closure was carried out with the epoxy resin constituent for the closures according to claim 1 to 5.

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[Field of the Invention]

[0001]

This invention relates to electronic-parts equipment equipped with the epoxy resin constituent for the closures excellent in the appearance on laser mark nature, an electrical property, a moldability, dependability, and the front face of a package, and the component by which the closure was carried out by this.

[Background of the Invention]

[0002]

The miniaturization of electronic-parts equipments, such as expansion of occupied volume [ as opposed to / semiconductor devices /, such as IC and LSI, / the package of a component in improvement in the degree of integration of a component ] and a semiconductor device, thin-shape-izing, and many pin-ization are progressing. Furthermore, it corresponds to the miniaturization of electronic equipment, and thin shape-ization, and the surface mount method with which the mounting approach also makes high density assembly possible has spread quickly instead of a pin inserting type method. Consequently, when attaching electronic-parts equipment in a substrate,

electronic-parts equipment itself is put to the inside of a short time at the elevated temperature of a solder bath 200 degrees C or more. At this time, the moisture contained in a sealing agent evaporates and the vapor pressure generated here works as exfoliation stress in the interface of a sealing agent and an insertion of a component, a leadframe, etc., and between a sealing agent and an insertion, exfoliation occurs and it keeps in blistering and the crack of electronic-parts equipment very much in thin electronic-parts equipment especially.

Although the technique of making it LOC (Lead on Chip) structure, and raising adhesion with a sealing agent is used or it performs dimple processing, slit processing, etc. at the technique and the island rear face of a leadframe which use coat material for a component front face or the island rear face of a leadframe, and raise adhesion with a sealing agent as a preventive measure of blistering by such exfoliation reason, and a crack, the problem of inadequate \*\* has a raise in cost, and effectiveness. Furthermore, in the case of LOC structure, the irregular color on the front face of a package arises, and an appearance is spoiled.

Moreover, the distance between [ , such as between inner leads, between pads, and between wires, ] pitches is becoming narrow with the miniaturization of electronic-parts equipments, such as a semiconductor device, and the formation of many pins, and when conductive matter, such as carbon black contained in a sealing agent, exists as big coarse grain, the carbon black which is conductivity keeps very much in narrowing and a poor electrical property between inner leads, between pads, and between wires. for this reason, examination which uses organic dye, a pigment, etc. instead of carbon black as a coloring agent is made -- \*\*\*\* (patent reference 1 and patent reference 2 reference.) -- there were problems, such as a fall of YAG laser mark nature, a fall of dependability, and a raise in cost.

[Patent reference 1] JP,63-179921,A

[Patent reference 2] JP,11-60904,A

[Description of the Invention]

[Problem(s) to be Solved by the Invention]

[0003]

This invention aims at offering electronic parts equipment equipped with the epoxy resin constituent for the closures which was made in view of this situation , and was excellent in laser mark nature or an electrical property , and short [ by the conductive matter / poor ] did not occurred in electronic parts equipments , such as between pads and a semiconductor device with a narrow distance between wires , and was excellent in the appearance on a moldability , dependability , and the front face of a package , and the component which this closed .

[Means for Solving the Problem]

[0004]

In order to solve the above-mentioned technical problem, as a result of repeating examination wholeheartedly, by blending non-conductive carbon with the epoxy resin constituent for the closures, this invention persons find out that the above-mentioned purpose can be attained, and came to complete this invention.

Namely, this invention,

(1) The epoxy resin constituent for the closures containing the color as which the (A) epoxy resin, the (B) curing agent, (C) non-conductive carbon, (D) inorganic filler, and (E) organic dye are used as an indispensable component, and \*\* (E) organic dye is chosen from an azine system color, anthraquinone dye, a JISUAZO system color, a G minium system color, an aminium system color, a G MONIUMU system color, and phthalocyanine dye,

(2) The epoxy resin constituent for the closures of the above-mentioned (1) publication with which the (E) color contains phthalocyanine dye,

(3) The above (1) whose electric resistance of non-conductive carbon is 107ohms or more, or the epoxy resin constituent for the closures given in (2),

(4) Above-mentioned (1) - (3) whose content of non-conductive carbon is 0.1 - 10% of the weight of the whole resin constituent is the epoxy resin constituent for the closures of a publication either,

(5) non-conductive carbon -- either [ by which the front face is covered with either at least / above-

mentioned (1) - (4) ] an epoxy resin and phenol resin -- the epoxy resin constituent for the closures of a publication -- and

(6) The above (1) Electronic-parts equipment equipped with the component by which the closure was carried out to either of - (5) with the epoxy resin constituent for the closures of a publication It is alike and is related.

[Effect of the Invention]

[0005]

The epoxy resin constituent for the closures which becomes this invention has a good moldability, as shown in the example, and since the electronic-parts equipment which is excellent in the appearance on laser mark nature, an electrical property, dependability, and the front face of a package is obtained, the industrial value is size.

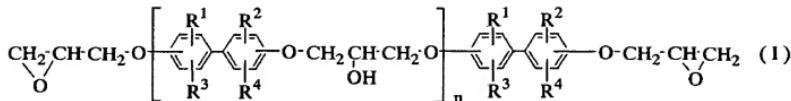
[Best Mode of Carrying Out the Invention]

[0006]

Although there is especially no limit as a (A) epoxy resin used in this invention For example, the phenol novolak mold epoxy resin currently generally used with the epoxy resin constituent for the closures, The epoxy resin which carried out epoxidation of the novolak resin compounded from phenols and aldehydes including polyglycidyl ether of o-cresol-formaldehyde novolac and bisphenol A novolak mold epoxy resin, Alkylation or an unsubstituted biphenol mold epoxy resin, alkylation, or an unsubstituted glycidyl ether mold epoxy resin, The glycidyl amine mold epoxy resin obtained by the reaction of polyamine and epichlorohydrin, such as isocyanuric acid, The epoxidation object of the copolycondensation resin of a biphenol mold epoxy resin, a dicyclopentadiene, phenols, and/or naphthols, The epoxy resin, phenol aralkyl resin which have a naphthalene ring, The epoxidation object of aralkyl mold phenol resin, such as naphthol aralkyl resin, the line oxidized and obtained by peroxy acids, such as a peracetic acid, in a trimethylol propane mold epoxy resin, a terpene modified epoxy resin, and olefin association -- an aliphatic series epoxy resin, an alicyclic group epoxy resin, etc. are mentioned, and these may be used independently or may be used combining two or more sorts.

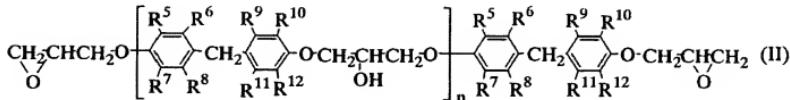
[0007]

Specifically, the compound shown by either the following general formula (I) - (IV) is mentioned.  
[Formula 1]



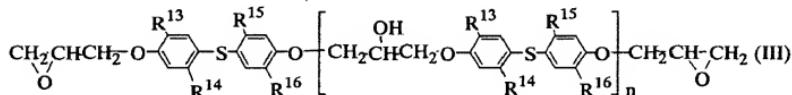
(Here, R1-R4 are chosen from the hydrocarbon group of the permutation of a hydrogen atom and carbon numbers 1-10, or unsubstituted monovalence, and even if all are the same, they may differ.)  
 $n$  shows the integer of 0-3.

[Formula 2]



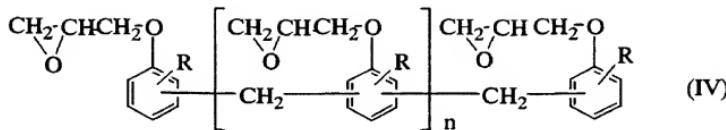
(Here, R5-R12 are chosen from the hydrocarbon group of the permutation of a hydrogen atom and carbon numbers 1-10, or unsubstituted monovalence, and even if all are the same, they may differ.) n shows the integer of 0-3.

[Formula 3]



(Here, R13-R16 are chosen from the hydrocarbon group of the permutation of hydrogen and carbon numbers 1-10, or unsubstituted monovalence, and even if altogether the same, they may differ.) n shows the integer of 0-10.

[Formula 4]



(Here, R is chosen from the hydrocarbon group of the permutation of hydrogen and carbon numbers 1-10, or unsubstituted monovalence, and n shows the integer of 0-10.)

[0008]

R1-R4 in the above-mentioned general formula (I) A hydrogen atom, a methyl group, an ethyl group, Alkyl groups, such as a propyl group, butyl, an isopropyl group, and an isobutyl radical, Alkoxy groups, such as a methoxy group, an ethoxy radical, a propoxy group, and a butoxy radical, Aralkyl radicals, such as aryl groups, such as a phenyl group, a tolyl group, and a xylyl group, benzyl, and a phenethyl radical, Although chosen out of the hydrocarbon group of the permutation of the carbon numbers 1-10, such as amino-group permutation alkyl groups, such as alkoxyalkyl groups, such as methoxymethyl radical and an ethoxymethyl radical methoxy ethyl group, an aminomethyl radical, and an aminoethyl radical, or unsubstituted monovalence, a hydrogen atom or a methyl group is desirable especially. As a biphenyl mold epoxy resin shown by the above-mentioned general formula (I) 4, a 4'-bis(2, 3-epoxy propoxy) biphenyl or 4, the 4'-screw (2, 3-epoxy propoxy) -3, 3', 5, the epoxy resin that uses a 5'-tetramethyl biphenyl as a principal component, Epichlorohydrin, 4 and 4'-biphenol or 4, the epoxy resin that 4'-(3, 3', 5, 5'-tetramethyl) biphenol is made to react, and is obtained are mentioned. Especially, 4, the 4'-screw (2, 3-epoxy propoxy) -3, 3', 5, and the epoxy resin that uses a 5'-tetramethyl biphenyl as a principal component are more desirable.

[0009]

R5-R12 in the above-mentioned general formula (II) A hydrogen atom, a methyl group, an ethyl group, Alkyl groups, such as a propyl group, butyl, an isopropyl group, and an isobutyl radical, Alkoxy groups, such as a methoxy group, an ethoxy radical, a propoxy group, and a butoxy radical, Aralkyl radicals, such as aryl groups, such as a phenyl group, a tolyl group, and a xylyl group, benzyl, and a phenethyl radical, Although chosen out of the hydrocarbon group of the permutation of the carbon numbers 1-10, such as amino-group permutation alkyl groups, such as alkoxyalkyl groups, such as a methoxymethyl radical and an ethoxymethyl radical methoxy ethyl group, an aminomethyl radical, and an aminoethyl radical, or unsubstituted monovalence, a hydrogen atom or a methyl group is desirable especially. As a bisphenol female mold epoxy resin shown by the above-mentioned general formula (II), for example, R5, R7, R10, and R12 are methyl groups, R6, R8, R9, and R11 are hydrogen atoms, and ESLV-80XY (trade name by Nippon Steel chemistry incorporated company) which uses n=0 as a principal component is available as a commercial item. [0010]

R13-R16 in the above-mentioned general formula (III) A hydrogen atom, a methyl group, an ethyl group, a propyl group, an isopropyl group, n-butyl, Alkyl groups, such as t-butyl, an amyl group, and isobutyl, a methoxy group, Alkoxy groups, such as an ethoxy radical, a propoxy group, and a butoxy radical, a phenyl group, Aralkyl radicals, such as aryl groups, such as a tolyl group and a xylyl group, benzyl, and a phenethyl radical, Although chosen out of the hydrocarbon group of the permutation of the carbon numbers 1-10, such as amino-group permutation alkyl groups, such as alkoxyalkyl groups, such as a methoxymethyl radical and an ethoxymethyl radical methoxy ethyl group, an aminomethyl radical, and an aminoethyl radical, or unsubstituted monovalence Especially, an alkyl group is desirable and a methyl group or t-butyl is more desirable. The epoxidation object of the bis(2-methyl-4-hydroxy-5-t-butyl) thioether which uses as a principal component the compound R13 and whose R16 are t-butyls, R14 and whose R15 are methyl groups, and whose n is 0 from availability and a fluid viewpoint as a phenylsulfide mold epoxy resin shown by the above-mentioned general formula (III) is desirable.

[0011]

As R in the above-mentioned general formula (IV), for example, the hydrocarbon group of the permutation of the carbon numbers 1-10, such as alkenyl radicals, such as alkyl groups, such as hydrogen, a methyl group, an ethyl group, a propyl group, butyl, an isopropyl group, and t-butyl, a vinyl group, an allyl group, and a butenyl group, an alkyl halide radical, an amino-group permutation alkyl group, and a sulphydryl group permutation alkyl group, or unsubstituted monovalence is mentioned, especially, alkyl groups, such as a methyl group and an ethyl group, are desirable, and a methyl group is more desirable. As a novolak mold epoxy resin shown by the above-mentioned general formula (IV), a phenol novolak mold epoxy resin, polyglycidyl ether of o-cresol-form aldehyde novolac, etc. are mentioned, and polyglycidyl ether of o-cresol-form aldehyde novolac is desirable especially.

[0012]

Although there is especially no limit as a (B) curing agent used in this invention, it is what is generally used with the epoxy resin constituent for the closures, for example. A phenol, cresol, resorcinol, a catechol, bisphenol A, Phenols, such as Bisphenol F, and/or the alpha-naphthol, the beta-naphthol, Naphthols, such as dihydroxy naphthalene, and aldehydes, such as formaldehyde, under an acid catalyst Condensation on the resin which is made to carry out copolycondensation and is obtained, Aralkyl mold phenol resin, such as phenol aralkyl resin and naphthol aralkyl resin, etc. is mentioned, and these may be used independently or may be used combining two or more sorts. (B) As for a curing agent, it is desirable to be blended to the epoxy group equivalent of the above-mentioned (A) epoxy resin in the amount from which the phenolated water acid radical equivalent turns into 0.5-1.5Eq, and it is more preferably blended by 0.8-1.2Eq. If hardening [ of an epoxy resin ] becomes insufficient [ less than 0.5Eq ], there is an inclination for an electrical property to be inferior to the thermal resistance of a hardened material and a damp-proof list and it exceeds 1.5Eq, in order that a curing agent component may become superfluous and a lot of phenolic hydroxyl groups may remain into hardening resin, the inclination for moisture resistance to worsen

is in an electrical property list.

[0013]

It is desirable to blend a hardening accelerator with it, in order to promote the etherification reaction of an epoxy group and a phenolic hydroxyl group in the epoxy resin constituent for the closures of this invention. Although there will be especially no limit if it is the compound which has the catalyst function in which the reaction of an epoxy resin and a curing agent is promoted, as a hardening accelerator For example, 1, 8-diaza-bicyclo (5, 4, 0) undecene - 7, 1, 5-diaza-bicyclo (4, 3, 0) nonene, 5, 6-dibutylamino -1, 8-diaza-bicyclo (5, 4, 0) undecene -7, benzyl dimethylamine, Tertiary amine and these derivatives, such as triethanolamine, dimethylamino ethanol, and a tris (dimethyl aminomethyl) phenol, Imidazole derivatives and these derivatives, such as 2-methylimidazole, 2-phenylimidazole, and 2-phenyl-4-methylimidazole, Tributyl phosphine, methyl diphenylphosphine, triphenyl phosphine, In organic phosphines and these phosphines, such as diphenylphosphine and a phenyl phosphine, a maleic anhydride, The phosphorus compounds which have intramolecular polarization which comes to add a compound with pi bonding, such as a benzoquinone and a diazo phenylmethane, Tetra-phenyl phosphonium tetraphenylborate, triphenyl phosphine tetraphenyl borate, 2-ethyl-4-methylimidazole tetraphenyl borate, N-methyl tetra-phenyl phosphonium-tetraphenyl borate, and triphenyl phosphonium-triphenyl borane etc. is mentioned, and these may be used independently or may be used combining two or more sorts.

The loadings of a hardening accelerator have desirable 0.01 - 5 weight section to the epoxy resin 100 weight section, and it is more desirable to blend 0.1 - 3 weight section. When [ than 5 weight sections ] more [ a facilitatory effect is small when fewer than the 0.01 weight section, and ], it is in the inclination for preservation stability to worsen.

[0014]

although there is especially no limit as (C) non-conductive carbon used in this invention -- for example

Non-conductive polymer baking carbon which is made to calcinate a polymer and is obtained, Graft carbon which is made to carry out the graft of the polymer on the surface of carbon black, and is obtained,

The carbon endocyst filler obtained by covering the front face of carbon black with insulating inorganic substances, such as a silica,

Carbon black which performed surface preparation, such as oxidation treatment

\*\*\*\* is mentioned, and these may be used independently or may be used combining two or more sorts. Moreover, with an epoxy resin, phenol resin, etc., the coat of these non-conductive carbon can be carried out, and it can also be used.

Although there is especially no limit in the manufacturing method of non-conductive polymer baking carbon, the method of, for example, calcinating polymers, such as a sulfonic-acid polymer which has aromatic series, such as naphthalene, at 400-600 degrees C is mentioned.

Although there is especially no limit in the manufacturing method of graft carbon, the method of carrying out the irreversible addition reaction of the graft polymer which has a carboxyl group on front faces of carbon black, such as an epoxy group, a thioepoxy radical, an aziridine radical, an oxazoline radical, and an isocyanate radical, and the radical which has reactivity, for example to carbon black with heat is mentioned. Although there is especially no limit as a graft polymer used here, copolymerization of the unsaturated compound which has the above-mentioned reactant radical is carried out to it and a copolymerizable monomer, it is obtained, and silicone, such as these derivatives, such as polystyrene, a polyethylene glycol, poly oxazoline, a polypropylene glycol, a polymethyl methacrylate, acrylic resin, and a fluororesin, and poly dimethylsiloxane, a wax, etc. are mentioned, for example, these may be used independently or may be used combining two or more sorts. Although there is especially no constraint as carbon black used for graft carbon, that in which many acid functional groups on a front face, such as a carboxyl group which is a reacting point with a graft polymer, exist is desirable, and seven or less carbon black has more desirable pH. The rate of a graft of graft carbon has 30 desirable % of the weight or more from a non-conductive viewpoint, and its 50 % of the weight or more is more desirable. Here, the rate of a

graft puts the rate of the graft polymer occupied to the whole graft carbon.

Although there is especially no limit in the manufacturing method of a carbon endocyst filler, the approach of carrying out the deposition of the silica particle by making the front face of the carbon black distributed in the water solution hydrolyze a tetra-ethoxy silane, the method of adding a silica sol to the carbon black distributed in the water solution, and carrying out the deposition of the silica particle to it with a sol gel process, etc. are mentioned, for example. Although there is especially no limit considering the front face of carbon black as a wrap insulation inorganic substance, metallic oxides, such as alkaline-earth-metal silicate, such as alkaline-earth-metal carbonates, such as a calcium carbonate and a barium carbonate, a calcium silicate, and a magnesium silicate, ferrous oxide, a silica, an alumina, oxidation copper, cobalt oxide, and nickel oxide, cobalt carbonate, nickel carbonate, basic copper carbonate, an aluminum hydroxide, a direct black 154 calcium lake, etc. are mentioned, and these may be used independently or may be used combining two or more sorts, for example. although there is especially no limit as carbon black used for a carbon endocyst filler -- water -- getting wet -- easy -- bulk -- a not high thing is desirable. The rate of carbon endocyst of a carbon endocyst filler has 70 or less desirable % of the weight from a non-conductive viewpoint, and its 50 or less % of the weight is more desirable. Here, the rate of carbon endocyst puts the rate of the carbon black occupied to the whole carbon endocyst filler.

As a manufacturing method of the carbon black which performed surface preparation, although there is especially no limit, the approach of carrying out surface preparation of the carbon black by the oxidizing method using an air-oxidation method, a nitric acid, nitrogen oxides, and oxidizing agents, such as mixed gas of air and ozone, the approach using a silane coupling agent, etc. is mentioned, for example.

[0015]

(C) The electric resistance of non-conductive carbon has desirable 107ohms or more, and is more desirable. [ of 109ohms or more ] Electric resistance is the comb mold electrode (35 micrometers of copper foil thickness) of the dimorphism which used glass fabric base material epoxy resin copper clad laminate MCL-E67 (trade name by Hitachi Chemical Co., Ltd.) as the substrate based on JIS Z3197. Between the line breadth of 0.3mm, and a line, using 0.3mm and 0.95mm of substrate thickness, powder sample 0.5g is fixed with a cellophane tape on wiring, and it asks by measuring the resistance of 1 minute after by direct-current 500V with a ohm-meter (TR8601 by Takeda Riken Industries).

(C) The mean particle diameter of non-conductive carbon has desirable 0.3-50 micrometers, and its 0.3-30 micrometers are more desirable. From a viewpoint of laser mark nature and a moldability, 0.3-10 micrometers is desirable and 0.3-5 micrometers is more desirable.

(C) The content of non-conductive carbon has 0.1 - 10 desirable % of the weight to the whole epoxy resin constituent for the closures, its 0.3 - 8 % of the weight is more desirable, and its 0.5 - 4 % of the weight is still more desirable. At less than 0.1 % of the weight, when the appearance on the front face of a package may be spoiled, or protection-from-light nature and laser mark nature may become inadequate and it exceeds 10 % of the weight, there is an inclination which becomes inadequate [ a moldability ].

[0016]

As a filler, it is required to use an inorganic filler from a viewpoint of hygroscopic reduction and the improvement in on the strength. Although there is especially no limit as a (D) inorganic filler used for this invention, it is what is generally used with the epoxy resin constituent for the closures, for example. Fused silica, a crystal silica, an alumina, zircon, a calcium silicate, a calcium carbonate, Single crystal fiber, such as fine particles, such as silicon carbide, boron nitride, beryllia, and a zirconia, or a bead which conglobated these, potassium titanate, silicon carbide, silicon nitride, and an alumina, a glass fiber, etc. are mentioned, and these may be used independently or may be used combining two or more sorts. Moreover, an aluminum hydroxide, \*\*\*\* zinc, etc. are mentioned as an inorganic filler with fire-resistant effectiveness, and these may be used independently or may be used together. Especially, the alumina from a viewpoint of high temperature conductivity [ fused silica / from a viewpoint of coefficient-of-linear-expansion

reduction ] is desirable, and the configuration of an inorganic filler has a desirable globular form from the point of the fluidity at the time of shaping, and metal mold abrasiveness.

(D) As loadings of an inorganic filler, 70 - 98 % of the weight is desirable to hygroscopicity, reduction of coefficient of linear expansion, the improvement in on the strength, and the viewpoint of solder thermal resistance to the whole epoxy resin constituent for the closures. It is 75 - 95% of the weight of the range more preferably.

[0017]

To the epoxy resin constituent for the closures of this invention, it is desirable to add a coupling agent from a viewpoint which plans the compatibility of an inorganic filler and a resinous principle. It is what especially a limit does not have as a coupling agent used, for example, is generally used with the epoxy resin constituent for the closures. Vinyl trichlorosilane, vinyltriethoxysilane, a vinyl tris (beta-methoxyethoxy) silane, Gamma-methacryloxypropyl trimethoxy silane, beta-(3, 4-epoxy dicyclohexyl) ethyltrimethoxysilane, Gamma-glycidoxypropytrimetoxysilane, vinyltriacetoxysilane, gamma-mercapto propyltrimethoxysilane, gamma-aminopropyl triethoxysilane, gamma-[bis(beta-hydroxyethyl)] amino pro BIRUTORIETOKISHISHIRAN, N-beta-(aminoethyl)-gamma-aminopropyl trimethoxysilane, gamma-(beta-aminoethyl) aminopropyl dimethoxymethylsilane, N-(trimethoxysilylpropyl) ethylenediamine, N-(dimethoxymethyl silyl isopropyl) ethylenediamine, Methyl trimetoxysilane, methyl triethoxysilane, N-beta-(N-vinylbenzyl aminoethyl)-gamma-aminopropyl trimethoxysilane, Gamma-chloropropyltrimetoxysilane, a hexamethyl disilane, gamma-ANIRINO propyltrimethoxysilane, Silane system coupling agents, such as vinyltrimetoxysilane and gamma-mercapto propylmethyl dimethoxysilane, Isopropylisostearoyl titanate, isopropyl tris (diocetylpyrophosphate) titanate, Isopropanal PIRUTORI (N-aminoethyl aminoethyl) titanate, Tetra-octyl bis(JITORIDESHIRO) sill phosphite titanate, tetrapod (2 and 2-diaryl oxy methyl-1-butyl) bis(JITORIDESHIRO) phosphite titanate, Bis(diocetylpyrophosphate) oxy-acetate titanate, Bis(diocetylpyrophosphate) ethylene titanate, isopropyl octanoyl titanate, Isopropylidimethyl acryloyltitanate, isopropyl tridecyl benzenesulphonyl titanate, Isopropyl ISOSUTEAROIRUJI acrylic titanate, isopropyl PIRUTORI (diocetyl phosphate) titanate, Titanate system coupling agents, such as isopropyl TORIKUMIRU phenyl titanate and tetra-isopropyl bis(diocetyl phosphite) titanate, etc. are mentioned, and these may be used independently or may be used combining two or more sorts.

[0018]

As other additives, if needed, it is independent respectively or ion trapper, such as reforming material, such as release agents, such as a higher fatty acid, for example, carnauba wax, a polyethylene system wax, etc., silicone oil, and silicone rubber, a hydrotalcite, and an antimony-bismuth, etc. can be blended combining two or more sorts. moreover, to the epoxy resin constituent for the closures of this invention If it is within the limits by which the effectiveness of this invention is attained, in addition to non-conductive carbon An azine system color, anthraquinone dye, a JISUAZO system color, a G minium system color, An aminium system color, a G MONIUMU system color, Cr complex, Fe complex, Co complex, coloring agents, such as metallic oxides, such as metallic compounds, such as nickel complex, and Fe, Cu, nickel, and aluminum, Mg, Fe, a mica, a near-infrared absorber, phthalocyanine pigment, phthalocyanine dye, and carbon black, -- one sort -- or two or more sorts can be used together. It is desirable to use phthalocyanine dye together from the point of laser mark nature, a fluidity, and hardenability especially. In using conductive particles, such as carbon black, together, in order to attain the purpose of this invention, 120cm thing 3/100g or less has [ the specific surface area which is excellent in dispersibility ] desirable DBP oil absorption below 130m<sup>2</sup>/g, and 80cm thing 3/100g or less has [ specific surface area ] more desirable DBP oil absorption below 100m<sup>2</sup>/g.

[0019]

If distributed mixing of each above-mentioned raw material can be carried out at homogeneity, no matter what technique it may use, it can prepare, but as a general approach, after the epoxy resin constituent for the closures of this invention mixes the raw material of predetermined loadings enough by a mixer etc., it can be kneaded with a hot calender roll, an extruder, etc., and can

mention the approach of cooling and pulverizing. It will be easy to use if it tablet-izes by a dimension and weight which suit a process condition.

[0020]

The epoxy resin constituent for the closures of this invention is excellent in laser mark nature and an electrical property, and is excellent in a moldability, dependability, and skeleton prevention, and can be used suitable for the closures, such as IC and LSI.

As electronic-parts equipment equipped with the component closed with the epoxy resin constituent for the closures obtained by this invention, components, such as passive elements, such as active elements, such as a semiconductor chip, a transistor, diode, and a thyristor, a capacitor, a resistor, and a coil, are carried in supporter material, such as a leadframe, a tape career [ finishing / wiring ], a patchboard, glass, and a silicon wafer, and the electronic-parts equipment which closed the required part with the epoxy resin constituent for the closures of this invention is mentioned. As such electronic-parts equipment, a semiconductor device is fixed on a leadframe, for example.

After connecting the terminal area and the lead sections of a component, such as a bonding pad, by wirebonding or the bump, Come to close by transfer molding etc. using the epoxy resin constituent for the closures of this invention. DIP (Dual Inline Package), PLCC (Plastic Leaded Chip Carrier), QFP (Quad Flat Package), SOP (Small Outline Package), SOJ (Small Outline J-lead package), TSOP (Thin Small OutlinePackage), The common plastic molded types IC, such as TQFP (Thin Quad Flat Package) TCP which closed the semiconductor chip connected to the tape carrier by the bump with the epoxy resin constituent for the closures of this invention (Tape Carrier Package), To wiring formed on a patchboard or glass, wirebonding, flip chip bonding, An active element and/or capacitors, such as a semiconductor chip connected with solder etc., a transistor, diode, and a thyristor, The COB (ChipOn Board) module which closed passive elements, such as a resistor and a coil, with the epoxy resin constituent for the closures of this invention, A component is carried in the front face of the organic substrate in which the terminal for patchboard connection was formed at the hybrid IC, the multi chip module, and the rear face. After connecting wiring formed in the component and the organic substrate of a bump or wirebonding, BGA (Ball Grid Array), CSP (Chip Size Package), etc. which closed the component with the epoxy resin constituent for the closures of this invention are mentioned. Moreover, the epoxy resin constituent for the closures of this invention can be effectively used also for a printed circuit board.

As an approach of closing electronic-parts equipment using the epoxy resin constituent for the closures obtained by this invention, although the low voltage transfer-molding method is the most common, approaches, such as injection shaping, compression molding, and casting, may be used.

[Example]

[0021]

Although an example explains this invention below at a detail, this invention is not limited to this. Examples 1-9, the examples 1-4 of a comparison

First, after carrying out preliminary mixing (dryblend) of the combination component shown in Table 1 and 2, with a 2 shaft roll (roll skin temperature of 80 degrees C), it kneaded for 10 minutes, cooling grinding was carried out, and the epoxy resin constituent for the closures of an example and the example of a comparison was manufactured.

[0022]

[Table 1]

表1 配合組成1

(配合量：重量部)

配合成分	実施例								
	1	2	3	4	5	6	7	8	9
ビフェニル型エポキシ樹脂	85.0	85.0	85.0	85.0	85.0	85.0	85.0	85.0	85.0
臭素化エポキシ樹脂	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0
アラルキル型フェノール樹脂	87.0	87.0	87.0	87.0	87.0	87.0	87.0	87.0	87.0
トリフェニルホスフィン	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
エポキシシラン	7.6	7.6	7.6	7.6	7.6	7.6	7.6	7.6	7.6
カルナバワックス	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
ポリエチレンワックス	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
非導電性カーボンA	55.0	-	-	20.0	20.0	10.0	-	-	30.0
非導電性カーボンB	-	60.0	-	-	30.0	20.0	-	-	-
非導電性カーボンC	-	-	65.0	-	-	30.0	-	-	-
非導電性カーボンD	-	-	-	-	-	-	50.0	-	-
非導電性カーボンE	-	-	-	-	-	-	-	30.0	-
カーボンブラックA	-	-	-	2.0	-	-	-	-	-
フタロシアニン系染料	-	-	-	-	-	-	-	-	1.0
三酸化アンチモン	12	12	12	12	12	12	12	12	12
溶融シリカ	1550	1550	1550	1550	1550	1550	1550	1550	1550

[0023]

[Table 2]

表2 配合組成2

(配合量：重量部)

配合成分	比較例			
	1	2	3	4
ビフェニル型エポキシ樹脂	85.0	85.0	85.0	85.0
臭素化エポキシ樹脂	15.0	15.0	15.0	15.0
アラルキル型フェノール樹脂	87.0	87.0	87.0	87.0
トリフェニルホスフィン	3.0	3.0	3.0	3.0
エポキシシラン	7.6	7.6	7.6	7.6
カルナバワックス	1.5	1.5	1.5	1.5
ポリエチレンワックス	1.5	1.5	1.5	0.5
カーボンブラックB	3.5	1.5	-	-
アジン系染料	-	10.0	35.0	-
斐母ー黒酸化鉄系染料	-	-	-	30.0
三酸化アンチモン	12	12	12	15
溶融シリカ	1550	1550	1550	1550

[0024]

In addition, Table 1 and the combination component given in Table 2 are as follows.

biphenyl mold epoxy resin: -- oil-ized shell epoxy incorporated company make -- trade name YX-4000H

bromination epoxy resin: -- the Tohto Kasei Co., Ltd. make -- trade name YDB-400

Aralkyl mold phenol resin: Trade name XL[ by Mitsui Chemicals, Inc. ]-225-3L

An epoxy silane: The trade name A-187 by Nippon Unicar, Inc.

Polyethylene wax: -- made in Clariant Japan, Inc. -- trade name PED-191

Non-conductive Carbon A:Mitsui Mining Co., Ltd. make -- trade name CB-3-500 (mean particle diameter of 3 micrometers, electric resistance of 109ohms)

Non-conductive Carbon B:Mitsui Mining Co., Ltd. make -- trade name CB-7-500 (mean particle diameter of 7 micrometers, electric resistance of 109ohms)

Non-conductive Carbon C:Mitsui Mining Co., Ltd. make -- trade name CB-10-500 (mean particle diameter of 10 micrometers, electric resistance of 109ohms)

Non-conductive carbon D: Graft carbon of 90 % of the weight of rates of a graft using styrene / acrylic graft polymer (NIPPON SHOKUBAI Make, electric resistance of 1010ohms)

Non-conductive carbon E: The carbon endocyst silica of 17 % of the weight of rates of carbon endocyst (the Suzuki fat-and-fatty-oil-industry incorporated company make, mean particle diameter of 7 micrometers, electric resistance of 109ohms)

Carbon black A: Carbon black (specific-surface-area 89m<sup>2</sup>/g and DBP oil absorption 60cm<sup>3</sup>/100g)

Carbon black B: Carbon black (specific-surface-area 140m<sup>2</sup>/g and DBP oil absorption

131cm<sup>3</sup>/100g)

Phthalocyanine dye: -- the Yamamoto Chemicals, INC. make -- trade name YKR-3080

Azine system color: Trade name spirit black 920 by Sumitomo Chemical Co., Ltd.

Mica-black-oxide-of-iron system color: The trade name LS 835 by Merck Japan, Inc.

Fused silica: Trade name S-CO by Micron, Inc.

[0025]

Each trial which shows the epoxy resin constituent for the closures of an example and the example of a comparison below estimated. In addition, shaping of the epoxy resin constituent for the closures was performed using the transfer-molding machine on the die temperature of 180 degrees C, compacting pressure 6.9MPa, and the conditions for setting-time 90 seconds. Moreover, posture (posture) was performed at 175 degrees C for 6 hours.

(1) Spiral flow

The metal mold according to EMM 11-66 was set to the transfer-molding machine, the epoxy resin constituent for the closures was fabricated on the above-mentioned conditions, and flow distance (cm) was found.

(2) It is a degree of hardness at the time of heat.

Weld flash measurement metal mold (metal mold which prepared width of face of 5mm, the depth 50, 30, 20, and 10, and a 5 or 2-micrometer slit) was set to the transfer-molding machine, the epoxy resin constituent for the closures was fabricated on the above-mentioned conditions, and a part for a resin pocket was measured with the Shore hardness tester after [ of metal mold disconnection ] 10 seconds.

(3) Volume resistivity

Disk metal mold was set to the transfer-molding machine, after fabricating and carrying out posture of the epoxy resin constituent for the closures to a disk with a diameter [ of 100mm ], and a thickness of 3mm on the above-mentioned conditions, using the volume-resistivity meter, it measured at electrical-potential-difference 500V and 150 degrees C, and insulation was checked.

(4) Moisture resistance

The electronic-parts equipment used for moisture resistance was a plastic molded type semiconductor device of SOP28 pin of SOP-28 pin (dimension of 18x8.4x2.6mm), and a leadframe has a 9.6x5.1mm TEG chip (with the aluminum wiring 10 and 20-micron width of face, a gap 10 and 20 microns, and no passivation) by 42 alloy material (processing dimple), and produced by performing transfer molding and posture on the above-mentioned conditions. After absorbing moisture by 85 degrees C / 85RH% for 72 hours, after pretreatment for 240 degrees C / 10 seconds

(IR reflow), PCT processing (121 degree-C/0.2MPa) was carried out, the existence of an open circuit of wiring on a chip was checked, and the PCT processing time until an open-circuit package reaches to 50% among a test package estimated.

#### (5) Solder thermal resistance

The electronic-parts equipment used for solder thermal resistance was a plastic molded type semiconductor device (dimension of 20x14x2.0mm) of QFP-80 pin, and a leadframe has the chip size of 8x10mm by 42 alloy material (with no processing), and produced by performing transfer molding and postcure on the above-mentioned conditions.

Thus, after predetermined carried out time amount moisture absorption of the obtained plastic molded type semiconductor device by 85 degrees C / 85RH%, the crack initiation when performing processing for 240 degrees C / 10 seconds was observed, and moisture absorption time amount until an appearance crack occurs estimated solder thermal resistance.

#### (6) Laser mark nature

The electronic-parts equipment used for laser mark nature evaluation was a plastic molded type semiconductor device of QFP-54 pin, it is YAG laser marking equipment, printed the package front face on condition that the YAG laser wavelength of 1064nm, and laser power 5J, and evaluated mark nature by viewing.

#### (7) Electrical property

The electronic-parts equipment used for evaluation of an electrical property is a plastic molded type semiconductor device of LQFP(Lowprofile Quad Flat Package)176 pin, and the existence of leakage current estimated it.

#### (8) Whenever [ black ]

Disk metal mold was set to the transfer-molding machine, and whenever [ black ] measured it with the color difference meter, after the front face fabricated and carried out postcure of the epoxy resin constituent for the closures to the disk with a diameter [ of 100mm ], and a thickness of 2mm which is crepe on the above-mentioned conditions. Whenever [ black ] shows black, so that a value is small.

The above-mentioned test result is shown in Table 3 and 4.

[0026]

[Table 3]

表3 評価結果 1

項目	単位	実施例								
		1	2	3	4	5	6	7	8	9
スパイアルフロー	cm	84	89	94	99	97	94	81	84	94
熱時硬度	-	80	80	81	80	81	81	80	81	81
体積抵抗率	$\Omega \cdot \text{cm}$	$6 \times 10^{13}$	$7 \times 10^{13}$	$6 \times 10^{13}$	$1 \times 10^{13}$	$5 \times 10^{13}$	$4 \times 10^{13}$	$1 \times 10^{13}$	$5 \times 10^{13}$	$4 \times 10^{13}$
耐湿性	h	650	650	600	720	600	650	650	600	650
半田耐熱性	h	120	120	120	120	120	120	120	120	120
レーザーマーク性	-	良好								
電気特性(リーク電流の有無)	-	無	無	無	無	無	無	無	無	無
黒色度	-	26.4	28.9	29.5	26.5	27.6	28.7	28.2	28.3	27.9
総合判定	-	○	○	○	○	○	○	○	○	○

[0027]

[Table 4]

表4 評価結果2

項目	単位	比較例			
		1	2	3	4
スパイアルフロー	cm	97	89	64	76
熱時硬度	-	81	80	79	80
体積抵抗率	$\Omega \cdot \text{cm}$	$3 \times 10^{13}$	$9 \times 10^{11}$	$8 \times 10^{11}$	$8 \times 10^{10}$
耐湿性	h	650	310	240	600
半田耐熱性	h	120	96	48	96
レーザーマーク性	-	良好	良好	良好	良好
電気特性(リード電流の有無)	-	有	無	無	有
黒色度	-	27.8	28.2	28.5	28.2
総合判定	-	×	×	×	×

[0028]

As for the examples 1-4 of a comparison which do not use non-conductive carbon as a coloring agent of this invention, a satisfactory property is acquired by neither. That is, the example 1 of a comparison using conductive carbon black is inferior to an electrical property, and the examples 2 and 3 of a comparison using an azine system color are inferior to moisture resistance and solder thermal resistance. The example 4 of a comparison using a mica-black-oxide-of-iron system color is inferior to solder thermal resistance and an electrical property.

On the other hand, examples 1-9 are excellent in dependability and electrical properties, such as moisture resistance and solder thermal resistance, and laser mark nature and also whenever [ black ] are good.

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## TECHNICAL FIELD

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### [Field of the Invention]

[0001]

This invention relates to electronic-parts equipment equipped with the epoxy resin constituent for the closures excellent in the appearance on laser mark nature, an electrical property, a moldability, dependability, and the front face of a package, and the component by which the closure was carried out by this.

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## PRIOR ART

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### [Background of the Invention]

[0002]

The miniaturization of electronic-parts equipments, such as expansion of occupied volume [ as opposed to / semiconductor devices /, such as IC and LSI, / the package of a component in improvement in the degree of integration of a component ] and a semiconductor device, thin-shape-zing, and many pin-ization are progressing. Furthermore, it corresponds to the miniaturization of electronic equipment, and thin shape-ization, and the surface mount method with which the mounting approach also makes high density assembly possible has spread quickly instead of a pin inserting type method. Consequently, when attaching electronic-parts equipment in a substrate, electronic-parts equipment itself is put to the inside of a short time at the elevated temperature of a

solder bath 200 degrees C or more. At this time, the moisture contained in a sealing agent evaporates and the vapor pressure generated here works as exfoliation stress in the interface of a sealing agent and an insertion of a component, a leadframe, etc., and between a sealing agent and an insertion, exfoliation occurs and it keeps in blistering and the crack of electronic-parts equipment very much in thin electronic-parts equipment especially.

Although the technique of making it LOC (Lead on Chip) structure, and raising adhesion with a sealing agent is used or it performs dimple processing, slit processing, etc. at the technique and the island rear face of a leadframe which use coat material for a component front face or the island rear face of a leadframe, and raise adhesion with a sealing agent as a preventive measure of blistering by such exfoliation reason, and a crack, the problem of inadequate \*\* has a raise in cost, and effectiveness. Furthermore, in the case of LOC structure, the irregular color on the front face of a package arises, and an appearance is spoiled.

Moreover, the distance between [, such as between inner leads, between pads, and between wires, ] pitches is becoming narrow with the miniaturization of electronic-parts equipments, such as a semiconductor device, and the formation of many pins, and when conductive matter, such as carbon black contained in a sealing agent, exists as big coarse grain, the carbon black which is conductivity keeps very much in narrowing and a poor electrical property between inner leads, between pads, and between wires. for this reason, examination which uses organic dye, a pigment, etc. instead of carbon black as a coloring agent is made -- \*\*\*\* (patent reference 1 and patent reference 2 reference.) -- there were problems, such as a fall of YAG laser mark nature, a fall of dependability, and a raise in cost.

[Patent reference 1] JP,63-179921,A

[Patent reference 2] JP,11-60904,A

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## EFFECT OF THE INVENTION

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[Effect of the Invention]

[0005]

The epoxy resin constituent for the closures which becomes this invention has a good moldability, as shown in the example, and since the electronic-parts equipment which is excellent in the appearance on laser mark nature, an electrical property, dependability, and the front face of a package is obtained, the industrial value is size.

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## TECHNICAL PROBLEM

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[Problem(s) to be Solved by the Invention]

[0003]

This invention aims at offering electronic parts equipment equipped with the epoxy resin constituent for the closures which was made in view of this situation , and was excellent in laser mark nature or an electrical property , and short [ by the conductive matter / poor ] did not occurred in electronic parts equipments , such as between pads and a semiconductor device with a narrow distance between wires , and was excellent in the appearance on a moldability , dependability , and the front face of a package , and the component which this closed .

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## MEANS

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[Means for Solving the Problem]

[0004]

In order to solve the above-mentioned technical problem, as a result of repeating examination

wholeheartedly, by blending non-conductive carbon with the epoxy resin constituent for the closures, this invention persons find out that the above-mentioned purpose can be attained, and came to complete this invention.

Namely, this invention,

- (1) The epoxy resin constituent for the closures containing the color as which the (A) epoxy resin, the (B) curing agent, (C) non-conductive carbon, (D) inorganic filler, and (E) organic dye are used as an indispensable component, and \*\* (E) organic dye is chosen from an azine system color, anthraquinone dye, a JISUAZO system color, a G minium system color, an aminium system color, a G MONIUM system color, and phthalocyanine dye,
- (2) The epoxy resin constituent for the closures of the above-mentioned (1) publication with which the (E) color contains phthalocyanine dye,
- (3) The above (1) whose electric resistance of non-conductive carbon is 107ohms or more, or the epoxy resin constituent for the closures given in (2),
- (4) Above-mentioned (1) - (3) whose content of non-conductive carbon is 0.1 - 10% of the weight of the whole resin constituent is the epoxy resin constituent for the closures of a publication either,
- (5) non-conductive carbon -- either [ by which the front face is covered with either at least / above-mentioned (1) - (4) ] an epoxy resin and phenol resin -- the epoxy resin constituent for the closures of a publication -- and
- (6) The above (1) Electronic-parts equipment equipped with the component by which the closure was carried out to either of - (5) with the epoxy resin constituent for the closures of a publication It is alike and is related.

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## EXAMPLE

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[Example]

[0021]

Although an example explains this invention below at a detail, this invention is not limited to this. Examples 1-9, the examples 1-4 of a comparison

First, after carrying out preliminary mixing (dryblend) of the combination component shown in Table 1 and 2, with a 2 shaft roll (roll skin temperature of 80 degrees C), it kneaded for 10 minutes, cooling grinding was carried out, and the epoxy resin constituent for the closures of an example and the example of a comparison was manufactured.

[0022]

[Table 1]

表1 配合組成1

(配合量：重量部)

配合成分	実施例								
	1	2	3	4	5	6	7	8	9
ビフェニル型エポキシ樹脂	85.0	85.0	85.0	85.0	85.0	85.0	85.0	85.0	85.0
臭素化エポキシ樹脂	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0
アラルキル型フェノール樹脂	87.0	87.0	87.0	87.0	87.0	87.0	87.0	87.0	87.0
トリフェニルホスフィン	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
エポキシシラン	7.6	7.6	7.6	7.6	7.6	7.6	7.6	7.6	7.6
カルナバワックス	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
ポリエチレンワックス	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
非導電性カーボンA	55.0	-	-	20.0	20.0	10.0	-	-	30.0
非導電性カーボンB	-	60.0	-	-	30.0	20.0	-	-	-
非導電性カーボンC	-	-	65.0	-	-	30.0	-	-	-
非導電性カーボンD	-	-	-	-	-	-	50.0	-	-
非導電性カーボンE	-	-	-	-	-	-	-	30.0	-
カーボンブラックA	-	-	-	2.0	-	-	-	-	-
フタロシアニン系染料	-	-	-	-	-	-	-	-	1.0
三酸化アンチモン	12	12	12	12	12	12	12	12	12
溶融シリカ	1550	1550	1550	1550	1550	1550	1550	1550	1550

[0023]

[Table 2]

表2 配合組成2

(配合量：重量部)

配合成分	比較例			
	1	2	3	4
ビフェニル型エポキシ樹脂	85.0	85.0	85.0	85.0
臭素化エポキシ樹脂	15.0	15.0	15.0	15.0
アラルキル型フェノール樹脂	87.0	87.0	87.0	87.0
トリフェニルホスフィン	3.0	3.0	3.0	3.0
エポキシシラン	7.6	7.6	7.6	7.6
カルナバワックス	1.5	1.5	1.5	1.5
ポリエチレンワックス	1.5	1.5	1.5	0.5
カーボンブラックB	3.5	1.5	-	-
アジン系染料	-	10.0	35.0	-
雲母-黒酸化鉄系染料	-	-	-	30.0
三酸化アンチモン	12	12	12	15
溶融シリカ	1550	1550	1550	1550

[0024]

In addition, Table 1 and the combination component given in Table 2 are as follows.

biphenyl mold epoxy resin: -- oil-ized shell epoxy incorporated company make -- trade name YX-4000H

bromination epoxy resin: -- the Tohto Kasei Co., Ltd. make -- trade name YDB-400

Aralkyl mold phenol resin: Trade name XL[ by Mitsui Chemicals, Inc. ]-225-3L

An epoxy silane: The trade name A-187 by Nippon Unicar, Inc.

Polyethylene wax: -- made in Clariant Japan, Inc. -- trade name PED-191

Non-conductive Carbon A:Mitsui Mining Co., Ltd. make -- trade name CB-3-500 (mean particle diameter of 3 micrometers, electric resistance of 109ohms)

Non-conductive Carbon B:Mitsui Mining Co., Ltd. make -- trade name CB-7-500 (mean particle diameter of 7 micrometers, electric resistance of 109ohms)

Non-conductive Carbon C:Mitsui Mining Co., Ltd. make -- trade name CB-10-500 (mean particle diameter of 10 micrometers, electric resistance of 109ohms)

Non-conductive carbon D: Graft carbon of 90 % of the weight of rates of a graft using styrene / acrylic graft polymer (NIPPON SHOKUBAI Make, electric resistance of 1010ohms)

Non-conductive carbon E: The carbon endocyst silica of 17 % of the weight of rates of carbon endocyst (the Suzuki fat-and-fatty-oil-industry incorporated company make, mean particle diameter of 7 micrometers, electric resistance of 109ohms)

Carbon black A: Carbon black (specific-surface-area 89m<sup>2</sup>/g and DBP oil absorption 60cm<sup>3</sup>/100g)

Carbon black B: Carbon black (specific-surface-area 140m<sup>2</sup>/g and DBP oil absorption 131cm<sup>3</sup>/100g)

Phthalocyanine dye: -- the Yamamoto Chemicals, INC. make -- trade name YKR-3080

Azine system color: Trade name spirit black 920 by Sumitomo Chemical Co., Ltd.

Mica-black-oxide-of-iron system color: The trade name LS 835 by Merck Japan, Inc.

Fused silica: Trade name S-CO by Micron, Inc.

[0025]

Each trial which shows the epoxy resin constituent for the closures of an example and the example of a comparison below estimated. In addition, shaping of the epoxy resin constituent for the closures was performed using the transfer-molding machine on the die temperature of 180 degrees C, compacting pressure 6.9MPa, and the conditions for setting-time 90 seconds. Moreover, postcure (postcure) was performed at 175 degrees C for 6 hours.

(1) Spiral flow

The metal mold according to EMM 11-66 was set to the transfer-molding machine, the epoxy resin constituent for the closures was fabricated on the above-mentioned conditions, and flow distance (cm) was found.

(2) It is a degree of hardness at the time of heat.

Weld flash measurement metal mold (metal mold which prepared width of face of 5mm, the depth 50, 30, 20, and 10, and a 5 or 2-micrometer slit) was set to the transfer-molding machine, the epoxy resin constituent for the closures was fabricated on the above-mentioned conditions, and a part for a resin pocket was measured with the Shore hardness tester after [ of metal mold disconnection ] 10 seconds.

(3) Volume resistivity

Disk metal mold was set to the transfer-molding machine, after fabricating and carrying out postcure of the epoxy resin constituent for the closures to a disk with a diameter [ of 100mm ], and a thickness of 3mm on the above-mentioned conditions, using the volume-resistivity meter, it measured at electrical-potential-difference 500V and 150 degrees C, and insulation was checked.

(4) Moisture resistance

The electronic-parts equipment used for moisture resistance was a plastic molded type semiconductor device of SOP28 pin of SOP-28 pin (dimension of 18x8.4x2.6mm), and a leadframe has a 9.6x5.1mm TEG chip (with the aluminum wiring 10 and 20-micron width of face, a gap 10 and 20 microns, and no passivation) by 42 alloy material (processing dimple), and produced by performing transfer molding and postcure on the above-mentioned conditions. After absorbing moisture by 85 degrees C / 85RH% for 72 hours, after pretreatment for 240 degrees C / 10 seconds

(IR reflow), PCT processing (121 degree-C/0.2MPa) was carried out, the existence of an open circuit of wiring on a chip was checked, and the PCT processing time until an open-circuit package reaches to 50% among a test package estimated.

(5) Solder thermal resistance

The electronic-parts equipment used for solder thermal resistance was a plastic molded type semiconductor device (dimension of 20x14x2.0mm) of QFP-80 pin, and a leadframe has the chip size of 8x10mm by 42 alloy material (with no processing), and produced by performing transfer molding and posture on the above-mentioned conditions.

Thus, after predetermined carried out time amount moisture absorption of the obtained plastic molded type semiconductor device by 85 degrees C / 85RH%, the crack initiation when performing processing for 240 degrees C / 10 seconds was observed, and moisture absorption time amount until an appearance crack occurs estimated solder thermal resistance.

(6) Laser mark nature

The electronic-parts equipment used for laser mark nature evaluation was a plastic molded type semiconductor device of QFP-54 pin, it is YAG laser marking equipment, printed the package front face on condition that the YAG laser wavelength of 1064nm, and laser power 5J, and evaluated mark nature by viewing.

(7) Electrical property

The electronic-parts equipment used for evaluation of an electrical property is a plastic molded type semiconductor device of LQFP(Lowprofile Quad Flat Package)176 pin, and the existence of leakage current estimated it.

(8) Whenever [ black ]

Disk metal mold was set to the transfer-molding machine, and whenever [ black ] measured it with the color difference meter, after the front face fabricated and carried out posture of the epoxy resin constituent for the closures to the disk with a diameter [ of 100mm ], and a thickness of 2mm which is crepe on the above-mentioned conditions. Whenever [ black ] shows black, so that a value is small.

The above-mentioned test result is shown in Table 3 and 4.

[0026]

[Table 3]

表3 評価結果1

項目	単位	実施例								
		1	2	3	4	5	6	7	8	9
スパイアルフロー	cm	84	89	94	99	97	94	81	84	94
熱時硬度	-	80	80	81	80	81	81	80	81	81
体積抵抗率	$\Omega \cdot \text{cm}$	$6 \times 10^{13}$	$7 \times 10^{13}$	$6 \times 10^{13}$	$1 \times 10^{13}$	$5 \times 10^{13}$	$4 \times 10^{13}$	$1 \times 10^{13}$	$5 \times 10^{13}$	$4 \times 10^{13}$
耐湿性	h	650	650	600	720	600	650	650	600	650
半田耐熱性	h	120	120	120	120	120	120	120	120	120
レーザーマーク性	-	良好								
電気特性(リーク電流の有無)	-	無	無	無	無	無	無	無	無	無
黒色度	-	26.4	28.9	29.5	26.5	27.6	28.7	28.2	28.3	27.9
総合判定	-	○	○	○	○	○	○	○	○	○

[0027]

[Table 4]

表4 評価結果2

項目	単位	比較例			
		1	2	3	4
スパイアルフロー	cm	97	89	64	76
熱時硬度	-	81	80	79	80
体積抵抗率	$\Omega \cdot \text{cm}$	$3 \times 10^{13}$	$9 \times 10^{11}$	$8 \times 10^{11}$	$8 \times 10^{10}$
耐湿性	h	650	310	240	600
半田耐熱性	h	120	96	48	96
レーザーマーク性	-	良好	良好	良好	良好
電気特性(リーク電流の有無)	-	有	無	無	有
黒色度	-	27.8	28.2	28.5	28.2
総合判定	-	×	×	×	×

[0028]

As for the examples 1-4 of a comparison which do not use non-conductive carbon as a coloring agent of this invention, a satisfactory property is acquired by neither. That is, the example 1 of a comparison using conductive carbon black is inferior to an electrical property, and the examples 2 and 3 of a comparison using an azine system color are inferior to moisture resistance and solder thermal resistance. The example 4 of a comparison using a mica-black-oxide-of-iron system color is inferior to solder thermal resistance and an electrical property.

On the other hand, examples 1-9 are excellent in dependability and electrical properties, such as moisture resistance and solder thermal resistance, and laser mark nature and also whenever [ black ] are good.